

HIGH-FREQUENCY ULTRASONIC CHARACTERIZATION OF ZrO_2 LAYER THICKNESSES USING DIGITAL SPECTRAL ANALYSIS

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INTRODUCTION

Embrittlement of Zr-2.5%Nb pressure tubes from CANDU nuclear reactors has recently become a concern in the Canadian nuclear industry. While a full understanding of the mechanisms involved has not yet been achieved, it is known that hydrogen/deuterium, liberated through oxidization of the tube's inner surface, is absorbed into the metal where it eventually precipitates out as weak, brittle hydride platelets. Knowledge of the nature and extent of oxide formation may thus give indications about the degree of hydrogen uptake and embrittlement in the tubes. In particular, a reliable, nondestructive technique for determining the thickness and integrity of these oxide layers is needed. Theoretically, such an evaluation is possible using ultrasonic spectroscopy.

The use of ultrasound to characterize layered media is by no means a new concept. As examples, Paradis, Serruys and Saglio [1] accurately measured the thicknesses of 250 μm aluminum sheets and similar success was achieved by Haynes, Bell and McIntyre [2] for epoxy and magnetite layers (140 - 650 μm) on mild steel. Generally, these attempts have been limited to well-defined, homogenous layers having well known (or predictable) acoustic properties, uniform thickness, and smooth, distinct interfaces. In contrast, little has previously been learned about such properties in pressure tube oxides. These oxide layers are, in addition, much thinner (20 - 40 μm). This defines a thickness regime where even small irregularities in the layer may drastically affect its ultrasonic response; it also necessitates the use of high-frequency (75 - 100 MHz) broadband ultrasound, introducing sound generation and attenuation difficulties.

THEORETICAL

A natural first approach, and the approach adopted here is the use of the single layer theory for a normally incident beam developed by Kino [3]

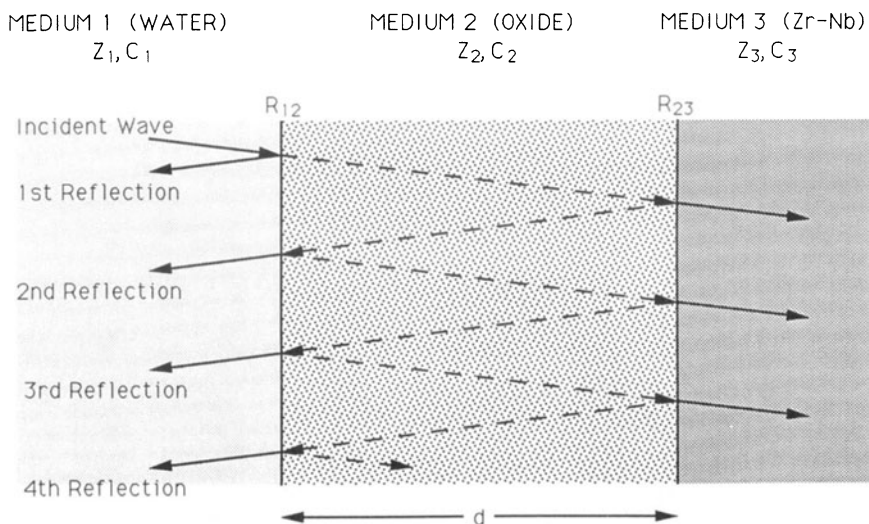


Fig. 1 Multiple reflection theory for single layer system. Skewed representation is for illustrative purposes only.

and Brekhovskikh [4] and illustrated in Fig. 1. For the very thin layer, the individual reflections become superimposed such that distinguishability is lost; simple time domain analysis is then unable to determine the time-of-flight of ultrasound within the layer or its "time-thickness". The reflection coefficient of the layer-substrate system can be calculated through solution of the continuous wave equations at the media boundaries.

$$|\hat{R}(f)|^2 = \frac{Z_2^2 (Z_3 - Z_1)^2 \cos^2 \beta d + (Z_2^2 - Z_1 Z_3)^2 \sin^2 \beta d}{Z_2^2 (Z_3 + Z_1)^2 \cos^2 \beta d + (Z_2^2 + Z_1 Z_3)^2 \sin^2 \beta d} \quad (1)$$

where: Z_1 is the acoustic impedance of the coupling medium (water),
 Z_2 is the acoustic impedance of the layer medium (oxide),
 Z_3 is the acoustic impedance of the substrate medium (Zr-2.5Nb),
 $\beta = \frac{\omega}{c} = \frac{2\pi}{\lambda}$ is the propagation constant in the layer,
 c is the acoustic velocity in the layer,
 d is the layer thickness,
 f is the frequency (ω is angular frequency) of the acoustic energy,
 λ is the wavelength of the acoustic energy.

The reflection coefficient magnitude is thus periodic in f as illustrated in Fig. 2, with minima occurring at frequencies

$$f = \frac{mc}{4d}, \quad m=1,3,5,\dots \quad \Rightarrow \quad \Delta f = \frac{c}{2d} \quad \Rightarrow \quad \Delta t = \frac{d}{c} = \frac{2}{\Delta f} \quad (2)$$

The index m can be eliminated by obtaining the separation, Δf , between dips and if c is known (or can be determined) then d can be calculated directly. Clearly then, the layer time-thickness, Δt , is just twice the inverse of Δf . Furthermore, the reflection coefficient minima are given by Eq. (3).

$$|\hat{R}(f)|_{\min} = \left| \frac{Z_2^2 - Z_1 Z_3}{Z_2^2 + Z_1 Z_3} \right| \quad (3)$$

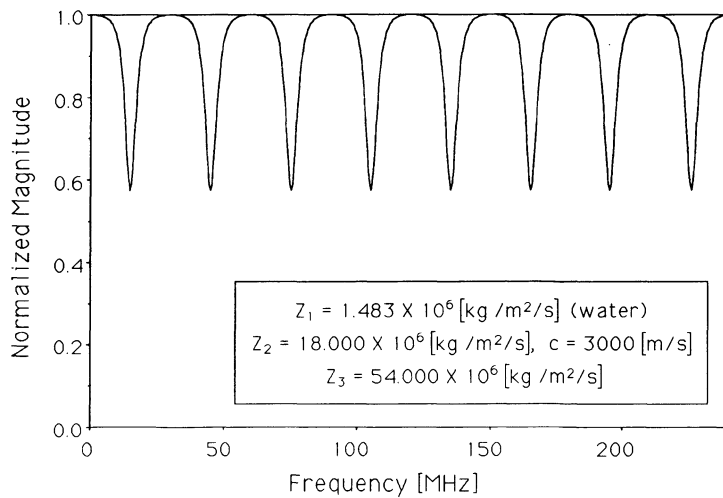


Fig. 2 Frequency dependent reflection coefficient for single layer system.

Since Z_1 and Z_3 are well known for this problem, Z_2 can thus be determined by measurement of $|\hat{R}(f)|_{\min}$. Thus, if bounds can be set *a priori* on either the velocity c or the density ρ of the layer then the thickness d can be determined, within like bounds.

This same result is obtained experimentally, using a *a priori* knowledge about the transducer response in the form of a reference signal from the unlayered system. The acquired reference and layered system signals are convolutions of the generated signal with reflection responses of each individual reflector system. Since the convolution operation becomes simple multiplication in the frequency domain, the dependence of acquired data on the input signal can be eliminated through division of the Fourier transform of the single-layer signal by that of the unlayered reference signal. The result is the reflection coefficient of Eq. (1) normalized by the frequency independent reflection coefficient of the simple flat reference reflector.

EXPERIMENTAL

Apparatus

Ultrasonic tests were carried out with Panametrics 75 MHz and 100 MHz broadband transducers, pulsed by a Panametrics 5601 pulser. Signals were digitized with a Tektronix 7854 digitizing oscilloscope, performing 100 sample averages to minimize the effects of random noise. Waveforms were then transferred to a Compaq 386/20 where all digital signal processing was performed using software developed at Ontario Hydro.

Digital Signal Processing

Waveforms were positioned to occupy approximately 1/4 of the time window and sampled at greater than twice the Nyquist frequency. They were gated and windowed by applying a narrow cosine half-cycle to either edge. A low-pass filter was then applied and the waveform decimated to provide the optimum sampling while minimizing aliasing effects. The resulting waveforms were four times zero-padded and the fast Fourier transforms were taken to obtain the complex spectrum of each.

The complex divisions of layered signals by a reference signal were performed using a Wiener type weighting in both the numerator and the denominator as in (4).

$$R(f) = \sqrt{\frac{\Phi_{\text{layer}}^2(f) + N_{\text{layer}}^2}{\Phi_{\text{ref}}^2(f) + N_{\text{ref}}^2}} \quad (4)$$

This provides easier interpretation of the spectral deconvolution than the conventional Wiener filter while still preventing division by zero problems. At frequencies where the magnitudes of both the signals are small, this division tends toward unity rather than zero as in conventional Wiener filtering. Since the information of the spectral deconvolution consists in dips from unity in the magnitude spectrum, the division employed here is less likely to result in misleading magnitude depressions.

Single-Layer Test Specimens

In order to evaluate the practicability of this analysis technique in the frequency and thickness regime representative of the expected oxides, several single-layer test specimens were prepared. These included brass, glass and plastic shims as well as polymer coatings on pressure tube metal, and were chosen to provide coherent, thin layers with very uniform intensive and extensive properties. Thicknesses were determined using time-thickness results using Eq. (2) and known bulk properties for all the samples, and impedance was determined from Eq. (3) for the polymer layers. Results from these experiments are presented in Fig. 3 with individual spectral deconvolutions in Fig. 4.

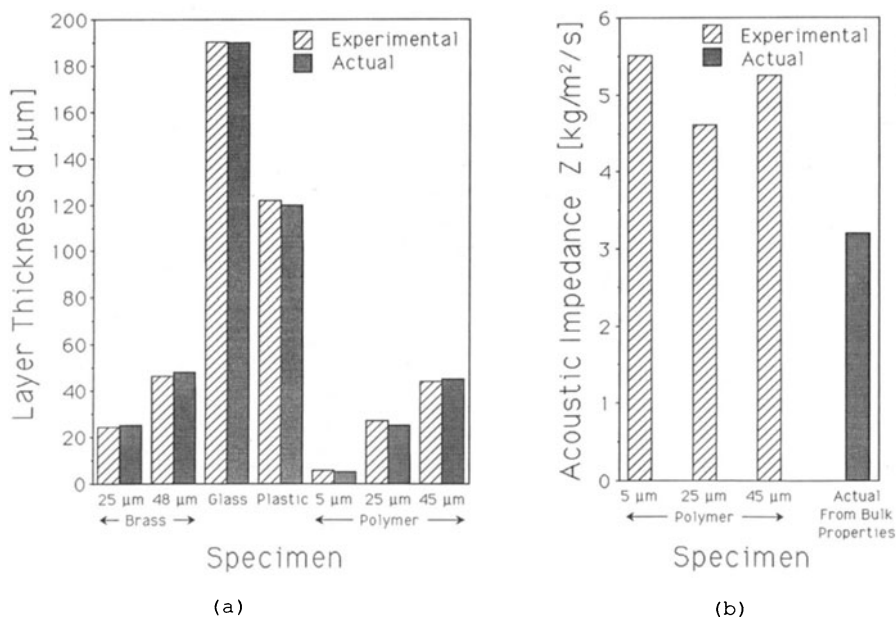


Fig. 3 Single-layer test specimen results: (a) ultrasonically determined vs. actual thicknesses and (b) ultrasonically determined vs. actual acoustic impedance of polymer layers.

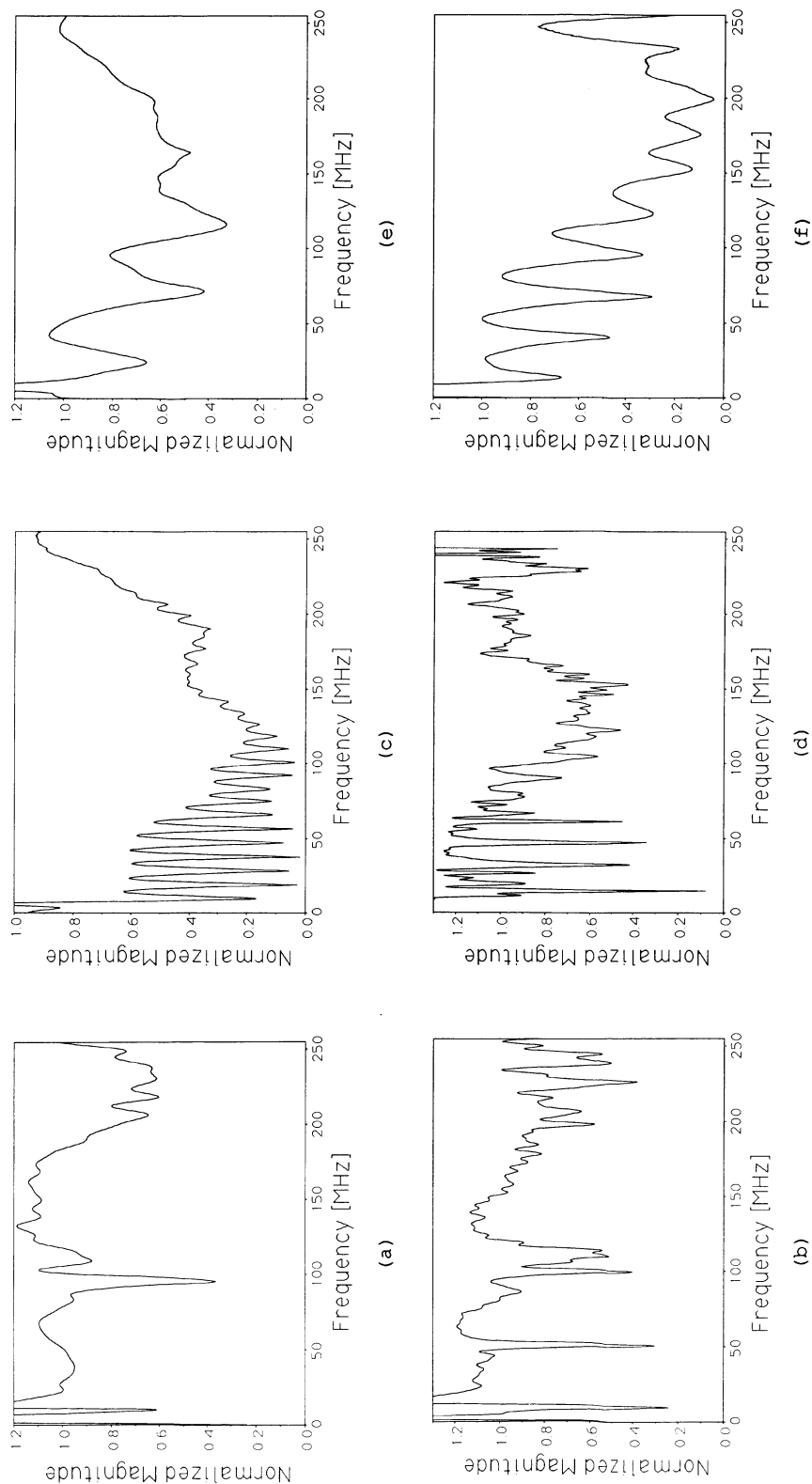


Fig. 4 Example spectral deconvolutions from single layer test samples: (a) 25 μm brass shim, (b) 48 μm brass shim, (c) 120 μm plastic slide, (d) 190 μm glass slide, (e) 25 μm polymer layer on Zr-2.5Nb and (f) 45 μm polymer layer on Zr-2.5Nb.

Oxide Specimens

The radioactivity of pressure tubes removed from operational reactors has precluded examination of oxide layers formed in service. In lieu of this, oxide specimens were prepared in an autoclave using sections of actual (unused) pressure tube. Since bulk properties are not known for the oxides, acoustic velocities were determined using both the location and the magnitude of the spectral dips correlated with the destructively determined thicknesses. These results are presented in Fig. 5. Typical individual spectral deconvolutions are presented in Fig. 6 outlining three basic groups, interpretable, null, and anomalous results.

DISCUSSION

It is clear from the individual test sample results of Fig. 3 that the theory and experiment are in good agreement, at least qualitatively, for this thickness regime. Dips occur as expected at regular intervals in the magnitude spectra beginning a half cycle from the frequency origin.

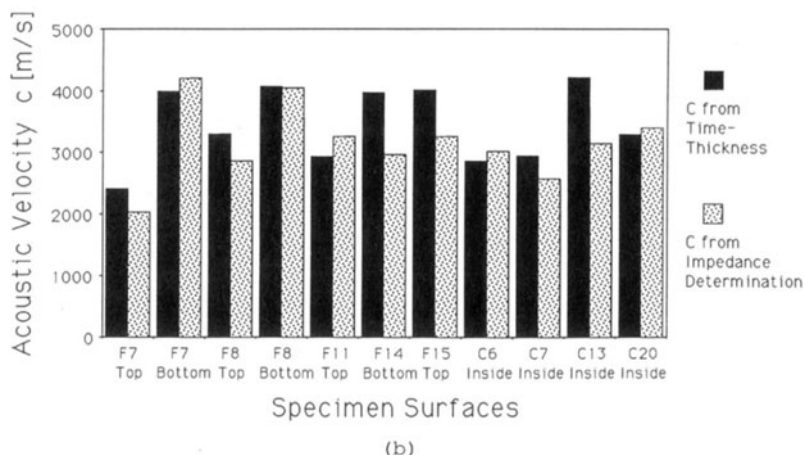
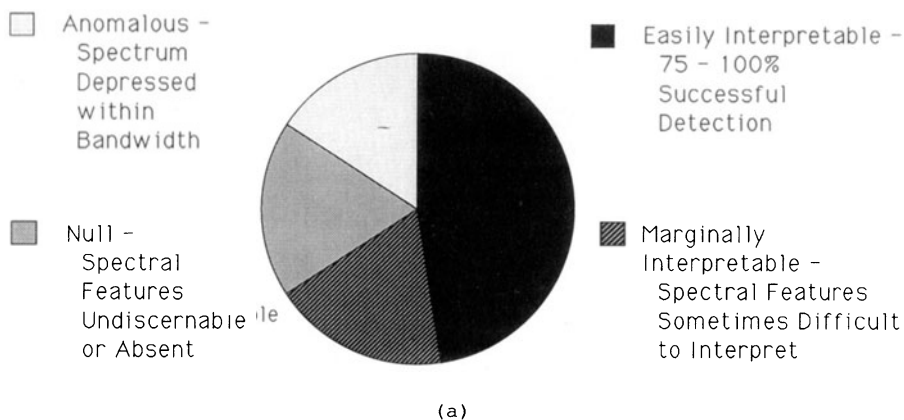


Fig. 5 Oxide experimental results: (a) Comparison of velocities from time-thickness and acoustic impedance results and (b) Detection efficiency of individual samples.

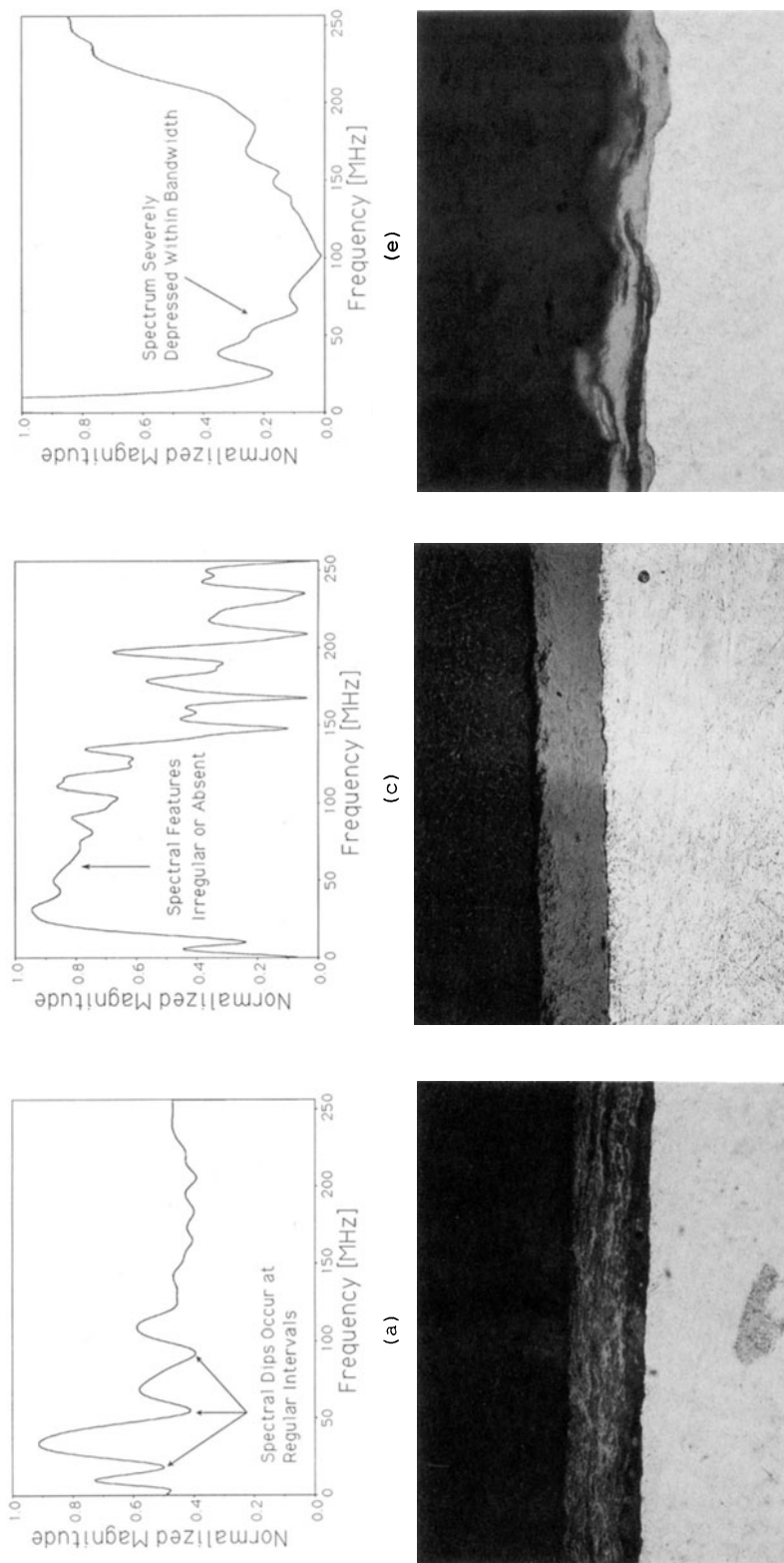


Fig. 6 Example spectral deconvolutions and corresponding oxides: (a) typical interpretable deconvolution and (b) corresponding oxide, (c) typical null result deconvolution and (d) corresponding oxide, (e) typical anomalous deconvolution and (f) corresponding oxide.

Closer examination in Fig. 4 shows that the time-thicknesses results are quite consistent and accurate. However, acoustic impedance determinations are inconsistent and inaccurate, and as such unreliable. It is quite likely that the magnitudes of the spectral dips are far more sensitive to layer irregularities than the frequencies at which they occur.

The example results in Fig. 6 indicate widely ranging characteristics of the oxide layers; correspondingly, there is also observed a wide range of behaviour in the resulting spectral divisions. Fig. 5(a) indicates that approximately half of the layers were readily detectable in terms of determining the time-thickness; Fig. 6(b) illustrates typical porous, flaky or layered substructure of the corresponding oxide layers. This suggests that the spectral analysis technique may be somewhat robust in detecting moderately nonuniform layers. In contrast, however, very dense consistent layers like that of Fig. 6(d) avoided detection. Presumably this is due to insufficient impedance mismatching between the layer and substrate; indeed the deconvolution magnitude spectra were virtually unaffected for these specimens, at most showing only slight hints of the presence of a layer. Depending on the properties of the actual in-service oxides, this method of layer measurement may be unsuitable for this reason. Very irregular layers as in Fig. 6(f) produced very depressed deconvolution spectra. This is not surprising; with such random and skewed geometry, one might expect much of the beam energy to be deflected at skew angles away from the transducer.

Fig. 5(b) attempts to compare the consistency of the results of examining both the location and depth of the spectral dips. By destructively measuring the thickness of each layer and by destructively determining an average density of the oxide, velocities resulting from both sets of measurements were calculated. The two sets of calculated velocities show modest agreement although results from the polymer layer experiments indicate that magnitudes of the spectral dips can be utilized with no better than marginal reliability. From sample to sample the values vary considerably, illustrating the wide variation in the oxide properties. However, numerical verification of actual thickness, just as verification of the applicability of this method to in-reactor oxides, depends entirely upon further quantification of the oxide properties.

REFERENCES

1. L. Paradis, R. Saglio and Y. Serruys, "Ultrasonic Signal Processing for Thickness Measurement and Detection of Near-Surface Defects", *Materials Evaluation* 44, Oct. 1986.
2. C. J. Bell, N. F. Haynes and P. J. McIntyre, "The Application of Broadband Ultrasonic Spectroscopy to the Study of Layered Media", *J. Acoust. Soc. Am.*, vol. 64, no. 1, pp 1645 - 51, Dec. 1978.
3. G. S. Kino, Acoustic Waves: Devices, Imaging and Analog Signal Processing, (Prentice-Hall, Englewood, New Jersey, 1987).
4. L. M. Brekhovskikh, Waves in Layered Media, 2nd ed., (Academic Press Inc, New York, 1980).